Requested Patent: GB1065568A

Title: POLYPROPYLENE BLENDS HAVING IMPROVED IMPACT STRENGTH ;

Abstracted Patent: GB1065568 ;

Publication Date: 1967-04-19 ;

Inventor(s): MARINAK MICHAEL JOSEPH ;

Applicant(s): DOW CHEMICAL CO ;

Application Number: GB19650053937 19651220 ;

Priority Number(s): US19640421664 19641228 ;

IPC Classification: ;

Equivalents: BE674416, FR1456359, NL6516008

ABSTRACT:

Compositions comprise a blend of (1) 70 to 94 weight per cent of isotactic polypropylene, (2) 1 to 25 weight per cent of an olefine polymer other than polypropylene or an ethylene/propylene copolymer and (3) 5 to 20 weight per cent of a normally solid ethylene/propylene copolymer. The olefine polymer may be a polymer or copolymer of ethylene, butene-1 and/or heptene-1. The compositions may be prepared by mixing or coprecipitation from a solvent such as isooctane or xylene, and may be moulded to form impact resistant boxes, tanks, luggage, car and refrigerator parts and battery boxes. Examples describe compositions of polypropylene, ethylene/propylene copolymers and polyethylene or polybutene-1.

1,065,568

PATENT SPECIFICATION

NO DRAWINGS

1,065,568

Inventor: MICHAEL JOSEPH MARINAK

Date of Application and filing Complete Specification: Dec. 20, 1965.

No. 53937/65.

Application made in United States of America (No. 421,664) on Dec. 28, 1964.

Complete Specification Published: April 19, 1967.

Index at acceptance:—C3 P(7C13B, 7C13C, 7D1A, 7D1C, 7D1X, 7K7)

Int. Cl.:—C 08 f 29/02

COMPLETE SPECIFICATION

Polypropylene Blends having improved Impact Strength

We, THE DOW CHEMICAL COMPANY, a Corporation organised and existing under the laws of the State of Delaware, United States of America, of Midland, County of Midland, State of Michigan, United States of America, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention relates to polypropylene compositions having improved impact strength and the method for making them. More particularly, this invention relates to a ternary mixture of polypropylene with an ethylene/propylene copolymer and another olefin polymer and its manufacture.

Polypropylene is a material which is highly regarded by the plastics industry for the reason that it possesses a number of desirable qualities, such as relatively low density, good flexibility and excellent heat resistance, and exhibits chemical inertness toward both aqueous and non-aqueous liquids. However, polypropylene has been found to be unduly susceptible to fracture by mechanical shock, especially at low temperatures. This deficiency has retarded the use of polypropylene in thin sectioned articles which may be struck a sharp blow at temperatures below freezing (0°C.), such as large boxes and tanks, luggage, automobile and refrigerator parts, battery boxes and so forth.

It is known that the incorporation of elastomers, such as polyisobutylene or amorphous ethylene/propylene copolymers will improve the impact strength of polypropylene. It is also known that these elastomers will improve the impact strength of either polyethylene or polypropylene blends at low temperatures, although they do not significantly improve the impact strength at room temperature. Additionally, the relatively large amount of elastomer necessary to effect the improvement adversely affects other valuable properties of the blend such as stiffness and tensile strength.

In accordance with the present invention it has been unexpectedly found that blending 5 to 20 per cent by weight of an ethylene-propylene copolymer with 70 to 94 per cent by weight of polypropylene along with 1 to 25 per cent by weight of an olefin polymer other than polypropylene or an ethylene/propoylene copolymer such as polyethylene, the resulting blends not only have improved impact strengths but have sufficient elastic modulus in tension to permit their use in hollow or thin sectioned articles, such as radio cabinets, containers, battery boxes and the like.

Polypropylene suitable for use in the compositions of this invention is a normally solid plastic material having molecular weights ranging from 5,000 to 500,000 especially 100,000 to 300,000. The usual commercially available grades which are substantially isotactic and suitable for molding, extrusion, coating, and the like shaping processes can be used.

The ethylene-propylene copolymers that may be incorporated in the polypropylene blends of the present invention are the normally solid, rubbery copolymers. These copolymers are materials known to the art and can be prepared by copolymerizing ethylene with propylene by any of several methods, such as the methods described in Belgian Patents 535,082, 538,782, 553,655, and 583,039 and U.S. Patents 2,700,663 and 2,726,231. Those copolymers containing from about 30 to about 70 per cent, most preferably from 40 to 60 mole per cent propylene residues are especially useful in preparing the blends of the present invention.

Olefin polymers which may be blended with polypropylene and ethylene/propylene copolymers to prepare the blends of the pre-

sent invention include polymers of ethylenically unsaturated monomers such as ethylene, butene-1, heptene-1, and copolymers thereof which may be of the low, intermediate or high density varieties or mixtures thereof having densities ranging from about 0.90 to about 0.97 gram/cubic centimeter; polyethylene having a density ranging from 0.940 to 0.960 gram/cubic centimeter is preferred.

The compounding of the polypropylene with the ethylene/propylene copolymers and the other olefin polymer can be done by malaxing, kneading, milling, or otherwise blending the materials together in any convenient order in the desired portions as by compounding the materials in an internal mixer such as a mixing extruder or a Banbury-type mixer, or on an external mixer such as an open-faced roll mill such as a rubber mill thereby to form intimately blended compositions. Further, these compositions can be prepared by coprecipitation from a neutral solvent such as isooctane or xylene.

Throughout the specification, parts and percentages are by weight unless otherwise specified. The example which follows illustrates the invention.

Example

Seventy parts of polypropylene, 10 parts of an ethylene/propylene copolymer rubber and 20 parts polyethylene were dry blended and then twice extruded in a mixing extruder at about 200°C.

The polypropylene was an isotactic polymer having a density of 0.90 grams per cubic centimeter, a melt index measured at 190°C. using a 2.16 kilogram weight of 0.79 decigram/minute, a tensile modulus of 120,000 pounds per square inch/8440 kg/cm² determined according to ASTM test designation D1708-59T, and a tensile yield stress of 4200 pounds per square inch/295 kg/cm².

The ethylene/propylene copolymer was ENJAY EPR 404, a rubber copolymer comprised of 40 to 46 per cent by weight ethylene, the remainder being propylene, having a specific gravity of 0.86 and a Mooney viscosity ML (1+8 min.) at 212°F./100°C. of 35—45.

The polyethylene was a high density polyethylene (0.964 gram/cubic centimeter) having a melt index of 0.23.

The blended material was formed by compression molding into test specimen sheets and notched bars were tested for impact resistance at ambient temperatures on an Izod type cantilever beam apparatus in accordance with ASTB test designation D256-56.

The impact resistance at below freezing temperatures of the blended material was determined using test specimen disks formed from the blended material by injection molding according to the Dart Drop Test ASTM test designation D1709-59T (modified).

Tensile yield stress and tensile modulus were determined in accordance with ASTM test designation D1708-59T.

For comparative purposes, blends of polypropylene-ethylene/propylene copolymer-polyethylene having different amounts of the polymeric materials inside and outside the ranges employed in the present invention and prepared from polymeric materials similar to those used above and blended in the same manner were also tested for impact resistance and tensile yield stress and tensile modulus.

The results of the tests were summarized in the table below.

TABLE

Sample No.	Composition of Blend: Poly-propylene (percent)	Composition of Blend: Ethylene-propylene Copolymer (percent)	Composition of Blend: Poly-ethylene (percent)	Tensile Yield (psi)/ kg/cm²	Tensile Modulus (psi)/ kg/cm²	Notched Izod Impact Strength at 73° F./ 23° C. Ft-lb/in/cm.kg/cm	Dart Drop at 0° F./ −18° C. Ft-lb/ kgm
1	70	10	20	2700/190	78,000/5485	14.6/78.9	50/6.91
2	75	15	10	2900/204	87,000/6116	14.0/78.3	35/4.83
3	85	10	5	3000/211	86,000/6045	12.3/67.0	20/2.76
4 (comparison)	87	3	10	3600/253	100,000/7030	1.2/6.5	<1.4/<0.19
5 (comparison)	91	3	6	3900/274	106,000/7450	1.6/8.7	<1.4/<0.19

By referring to the above Table, it is at once apparent that the polypropylene-ethylene/propylene copolymer-polyethylene blends of the present invention (sample numbers 1 through 3) have significantly improved impact resistance when compared to polypropylene-ethylene/propylene copolymer-polyethylene blends outside the scope of the present invention (sample numbers 4 and 5) and have sufficient tensile modulus to permit their use in thin sectioned articles manufactured therefrom.

In a similar manner the blending of other olefin polymers mentioned above such as polybutene-1 with polypropylene and ethylene/propylene copolymers in the proportions above specified causes a similar improvement in the impact resistance of the blends.

WHAT WE CLAIM IS:—

1. A composition comprising a blend of
(1) 70 to 94 per cent by weight of isotactic polypropylene,
(2) 1 to 25 per cent cent by weight of an olefin polymer other than polypropylene or an ethylene/propylene copolymer, and
(3) 5 to 20 per cent by weight of a normally solid ethylene/propylene copolymer.

2. A composition as claimed in Claim 1 wherein the olefin polymer (2) is polyethylene.

3. A composition as claimed in Claim 2, wherein the polyethylene has a density from 0.94 to 0.96 gram/cubic centimeters.

4. A composition as claimed in any one of Claims 1 to 3, wherein the ethylene/

propylene copolymer contains 40 to 60 mole per cent of a copolymerized ethylene, and from 60 to 40 mole per cent of copolymerized propylene.

5. A composition as claimed in any one of Claims 1 to 4, wherein the isotactic polypropylene has a molecular weight of 100,000 to 300,000.

6. A composition as claimed in any one of Claims 1, 4 or 5 wherein the olefin polymer (2) is a polymer of butene-1 or heptene-1.

7. The method of improving the impact strength of polypropylene which comprises blending with 70 to 94 per cent by weight polypropylene 1 to 25 per cent by weight of an olefin polymer other than polypropylene or an ethylene/propylene copolymer and 5 to 20 per cent by weight of a normally solid ethylene/propylene copolymer.

8. The method of Claim 7 wherein the olefin polymer is polyethylene.

9. The method of Claim 7 or 8, wherein the olefin polymer is a polyethylene having a density from 0.94 to 0.96 gram/cubic centimeter.

10. The method of any one of Claims 7 to 9 wherein the ethylene/propylene copolymer contains 40 to 60 mole per cent of copolymerized ethylene and from 60 to 40 mole per cent of copolymerized propylene.

11. The method of any one of Claims 7 to 10, wherein the isotactic polypropylene has a molecular weight of 100,000 to 300,000.

12. A polypropylene composition as claimed in Claim 1 substantially as described in the specific Example.

BOULT, WADE & TENNANT,
111 & 112, Hatton Garden,
London, E.C.1.
Chartered Patent Agents.
Agents for the Applicants.

Leamington Spa: Printed for Her Majesty's Stationery Office, by the Courier Press (Leamington) Ltd.—1967. Published by The Patent Office, 25 Southampton Buildings, London, W.C.2, from which copies may be obtained.